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UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 CFR 1.53(b))

Attorney Docket No.	12,150
First Inventor	David W. Warren
Title	THERMALLY-INTEGRATED LOW TEMPERATURE WATER-GAS SHIFT REACTOR APPARATUS AND PROCES
Express Mail Label No.	

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

ADDRESS TO: Assistant Commissioner for Patents
Box Patent Application
Washington, DC 20231

1. [X] Fee Transmittal Form (e.g., PTO/SB/17) *(Submit an original and a duplicate for fee processing)*
2. [X] Applicant claims small entity status. See 37 CFR 1.27.
3. [X] Specification [Total Pages 26] *(preferred arrangement set forth below)*
   - Descriptive title of the invention
   - Cross Reference to Related Applications
   - Statement Regarding Fed sponsored R & D
   - Reference to sequence listing, a table, or a computer program listing appendix
   - Background of the Invention
   - Brief Summary of the Invention
   - Brief Description of the Drawings *(if filed)*
   - Detailed Description
   - Claim(s)
   - Abstract of the Disclosure
4. [X] Drawing(s) *(35 U.S.C. 113)* [Total Sheets 4]
5. Oath or Declaration [Total Pages 2]
   a. [X] Newly executed (original or copy)
   b. [ ] Copy from a prior application (37 CFR 1.63 (d)) *(for continuation/divisional with Box 17 completed)*
      i. [ ] DELETION OF INVENTOR(S) Signed statement attached deleting inventor(s) named in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).
6. [ ] Application Data Sheet. See 37 CFR 1.76
7. [ ] CD-ROM or CD-R in duplicate, large table or Computer Program *(Appendix)*
8. Nucleotide and/or Amino Acid Sequence Submission *(if applicable, all necessary)*
   a. [ ] Computer Readable Form (CRF)
   b. Specification Sequence Listing on:
      i. ☐ CD-ROM or CD-R (2 copies); or
      ii. ☐ paper
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ACCOMPANYING APPLICATION PARTS

9. [X] Assignment Papers (cover sheet & document(s))
10. [ ] 37 CFR 3.73(b) Statement *(when there is an assignee)* [ ] Power of Attorney
11. [ ] English Translation Document *(if applicable)*
12. [X] Information Disclosure Statement (IDS)/PTO-1449 [X] Copies of IDS Citations
13. [ ] Preliminary Amendment
14. [X] Return Receipt Postcard (MPEP 503) *(Should be specifically itemized)*
15. [ ] Certified Copy of Priority Document(s) *(if foreign priority is claimed)*
16. [ ] Other: .....

17. If a CONTINUING APPLICATION, *check appropriate box, and supply the requisite information below and in a preliminary amendment, or in an Application Data Sheet under 37 CFR 1.76:*

☐ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No. _____/_____

Prior application information: Examiner _____ *Group / Art Unit.* _____

For CONTINUATION OR DIVISIONAL APPS only: The entire disclosure of the prior application, from which an oath or declaration is supplied under Box 5b, is considered a part of the disclosure of the accompanying continuation or divisional application and is hereby incorporated by reference. The incorporation can only be relied upon when a portion has been inadvertently omitted from the submitted application parts.

18. CORRESPONDENCE ADDRESS

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Signature	[signature]	Date	11-8-00

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FEE TRANSMITTAL for FY 2001

Patent fees are subject to annual revision.

TOTAL AMOUNT OF PAYMENT ($) 395.00

Complete if Known	
Application Number	
Filing Date	
First Named Inventor	David W. Warren et al
Examiner Name	
Group Art Unit	
Attorney Docket No.	12,150

METHOD OF PAYMENT

1. [X] The Commissioner is hereby authorized to charge indicated fees and credit any overpayments to.
   Deposit Account Number: 08-0118
   Deposit Account Name: William W. Haefliger
   [ ] Charge Any Additional Fee Required Under 37 CFR 1.16 and 1.17
   [X] Applicant claims small entity status. See 37 CFR 1.27
2. [X] **Payment Enclosed:**
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FEE CALCULATION

1. BASIC FILING FEE

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
101	710	201	355	Utility filing fee	355.
106	320	206	160	Design filing fee	
107	490	207	245	Plant filing fee	
108	710	208	355	Reissue filing fee	
114	150	214	75	Provisional filing fee	

SUBTOTAL (1) ($) 355.

2. EXTRA CLAIM FEES

		Extra Claims		Fee from below	Fee Paid
Total Claims	14	-20** =		X	=
Independent Claims	1	- 3** =		X	=
Multiple Dependent					=

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description
103	18	203	9	Claims in excess of 20
102	80	202	40	Independent claims in excess of 3
104	270	204	135	Multiple dependent claim, if not paid
109	80	209	40	** Reissue independent claims over original patent
110	18	210	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) ($)

**or number previously paid, if greater, For Reissues, see above

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
105	130	205	65	Surcharge - late filing fee or oath	
127	50	227	25	Surcharge - late provisional filing fee or cover sheet	
139	130	139	130	Non-English specification	
147	2,520	147	2,520	For filing a request for *ex parte* reexamination	
112	920*	112	920*	Requesting publication of SIR prior to Examiner action	
113	1,840*	113	1,840*	Requesting publication of SIR after Examiner action	
115	110	215	55	Extension for reply within first month	
116	390	216	195	Extension for reply within second month	
117	890	217	445	Extension for reply within third month	
118	1,390	218	695	Extension for reply within fourth month	
128	1,890	228	945	Extension for reply within fifth month	
119	310	219	155	Notice of Appeal	
120	310	220	155	Filing a brief in support of an appeal	
121	270	221	135	Request for oral hearing	
138	1,510	138	1,510	Petition to institute a public use proceeding	
140	110	240	55	Petition to revive - unavoidable	
141	1,240	241	620	Petition to revive - unintentional	
142	1,240	242	620	Utility issue fee (or reissue)	
143	440	243	220	Design issue fee	
144	600	244	300	Plant issue fee	
122	130	122	130	Petitions to the Commissioner	
123	50	123	50	Petitions related to provisional applications	
126	240	126	240	Submission of Information Disclosure Stmt	
581	40	581	40	Recording each patent assignment per property (times number of properties)	40.
146	710	246	355	Filing a submission after final rejection (37 CFR § 1.129(a))	
149	710	249	355	For each additional invention to be examined (37 CFR § 1 129(b))	
179	710	279	355	Request for Continued Examination (RCE)	
169	900	169	900	Request for expedited examination of a design application	

Other fee (specify) _____

* Reduced by Basic Filing Fee Paid

SUBTOTAL (3) ($) 40.

SUBMITTED BY

Name (Print/Type)	William W. Haefliger	Registration No. (Attorney/Agent)	17,120	Telephone	(323) 684-2707
Signature				Date	11-8-00

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TO WHOM IT MAY CONCERN:

BE IT KNOWN THAT WE, DAVID W. WARREN, a citizen of the United States of America, residing in Van Nuys, in the County of Los Angeles, and MICHAEL B. DONAHUE, a citizen of the United States of America, residing in La Verne, in the County of Los Angeles, both in the State of California, have invented a new and useful improvement in

THERMALLY-INTEGRATED LOW TEMPERATURE WATER-GAS SHIFT REACTOR APPARATUS AND PROCESS

BACKGROUND OF THE INVENTION

This invention relates to a process and an apparatus for producing hydrogen for application to fuel cell electric generators.

Catalytic reaction apparatus and processes for converting hydrocarbon feedstocks to useful industrial gases, such as hydrogen, is well known in the art. Proton exchange membrane (PEM) fuel cells have emerged as a viable option for the production of disbursed electrical power, typically in the range of 2-50 k W, for use in residential and small commercial applications. PEM fuel cells generate electricity by the electrochemical reaction between hydrogen and oxygen.

While oxygen is readily available from ambient air, hydrogen must be produced from commercially available fuels, such a natural gas or propane, using methods such as steam reforming. Steam reforming is a process that involves a high temperature catalytic reaction between a hydrocarbon and steam to form a hydrogen-rich product gas, commonly referred to as reformate, that contains significant quantities of carbon monoxide.

Since PEM fuel cells have a low tolerance to carbon monoxide, the concentration of carbon monoxide in the reformate must be reduced using a catalytic reaction step known as the water-gas shift reaction. Following the water-gas shift reaction, the concentration of carbon monoxide in the reformate is further reduced to concentrations typically less than 10 ppm using a selective oxidation reaction, also referred to as preferential oxidation or PROX. The combination of processes that convert commercial fuels to a reformate suitable for use in a fuel cell is commonly referred to as a fuel processor.

As an illustration, Table 1 summarizes the reaction steps of a fuel processor designed to produce a hydrogen-rich gas stream suitable for use in a PEM fuel cell.

Table 1. PEM fuel processor reaction steps

1.	$CH_4 + H_2O = CO + H_2$	Steam reforming
2.	$CO + H_2O = CO_2 + H_2$	Water-gas shift
3.	$CO + \frac{1}{2}O_2 = CO_2$	Selective oxidation

In typical industrial practice, the water-gas shift reaction is conducted in two separate adiabatic reactors operating at two different temperature

regimes. The first reactor, known as the high temperature shift reactor, operates at inlet temperatures typically ranging from about 550°F to 650°F. The second reactor, known as the low temperature shift reactor, operates at an inlet temperature typically ranging from about 380°F to 450°F. The combination of the two sequential water-gas shift reactions typically reduces the concentration of carbon monoxide in the reformate to less than 1.0 volume percent.

The use of a low temperature shift reactor is beneficial because the water-gas shift reaction is thermodynamically favored at lower temperatures. However, a high temperature shift reactor is generally required to limit the amount of heat that is released in the low temperature shift reactor.

Conventional low temperature shift catalysts comprise a mixture of copper and zinc that are supported on a ceramic carrier. These catalysts promote the water-gas shift reaction at lower temperature, but lose activity if they are exposed to excessively high temperatures due to sintering of the active metals. Therefore, it is generally desirable to limit the maximum temperature of the low temperature shift catalyst to about 500°F in order to achieve long catalyst life.

The water-gas shift reaction releases approximately 9837 calories per gram-mole of carbon monoxide that is consumed. If the water-gas shift reaction were conducted using a single adiabatic low temperature shift reactor, the heat release would result in a temperature increase across the catalyst bed that would exceed the desirable temperature limit for conventional low temperature shift reactors. Furthermore, the high exit temperature from the water-gas shift reactor would be thermodynamically less favorable for achieving high conversions of carbon monoxide.

There is need to minimize the number of reactors and heat exchangers that are needed to achieve the objective of high conversion of carbon monoxide for PEM fuel cell applications, in order to reduce the size, cost and complexity of the fuel processor. Therefore, it is desirable to conduct the water-gas shift reaction using a single reactor vessel that is maintained within acceptable operating temperature limits by controlling heat removal from the reactor.

The steam reforming reaction requires large quantities of steam for the conversion of hydrocarbon to reformate. It is desirable to recover the heat released from the water-gas shift reaction for the purpose of generating steam in order to improve the

thermal efficiency of the fuel processor. The present invention achieves the objective of temperature control and heat recovery by integrating a lower temperature shift reactor within a steam generator that contains water boiling at a temperature range of about 360°F to 400°F, corresponding to a boiler pressure of about 153 psia to 247 psia.

Because the vessel walls of the lower temperature shift reactor are in heat transfer communication with boiling water, the heat released from the water-gas shift reaction is effectively removed to control the temperature in the catalyst bed within the desired operating temperature range. Furthermore, the heat released from the water-gas shift reaction is beneficially recovered to generate steam that is used in the process. Finally, the steam generator provides a convenient source of heat for heating the catalyst bed during start-up.

U.S. Patent 6,086,840 describes a process for making ammonia that mentions use of an isothermal shift reactor that includes heat exchange tubes extending within a vessel packed with catalyst. The heat exchange tubes contain a boiling fluid to remove heat from the catalyst bed.

SUMMARY OF THE INVENTION

It is the general object of this invention to provide a novel catalytic reaction apparatus and process for the reduction of carbon monoxide contained in hydrogen-rich gas streams, employing lower temperature shift reactor that is in thermal communication with a steam generator operating in a temperature range that is optimum for catalytic activity and thermodynamic conversion, and allows the recovery of useful waste heat to generate steam needed for a process to convert hydrocarbons feedstocks to useful gases, such as hydrogen. The subject invention is particularly well suited for the production of hydrogen for fuel cells having low tolerance to carbon monoxide.

Basically, the invention provides a thermally-integrated low temperature water-gas shift reactor for converting carbon monoxide in the presence of steam to form carbon dioxide and water comprising, in combination,

a) a waste-heat recovery steam generator for the beneficial recovery of exothermic reaction heat to generate steam that is used in a process for the

conversion of hydrocarbon feedstock into hydrogen-rich gases,

b) an outer annulus extending about said waste-heat steam generator,

c) a catalyst bed located within said outer region, and through which reformate gases flow,

d) the outer region being in heat transfer communication with the steam generator to maintain the catalyst bed within a predetermined temperature range for operation of a low temperature shift reaction.

These and other objects and advantages of the invention, as well as the details of an illustrative embodiment, will be more fully understood from the following specification and drawings, in which:

DRAWING DESCRIPTION

Fig. 1 is a flow diagram of a process that incorporates a thermally-integrated low temperature water-gas shift reactor;

Fig. 2 is a schematic showing of apparatus for testing a thermally-integrated low temperature water-gas shift apparatus;

Fig. 3a is a schematic showing of a thermally-integrated low temperature water-gas shift reactor apparatus;

Fig. 3b is a section taken on lines 3b-3b of Fig. 3a; and

Fig. 4 is a schematic showing of an alternate thermally-integrated low temperature water-gas shift reactor apparatus.

DETAILED DESCRIPTION

PROCESS AND APPARATUS

The process and apparatus as shown in Fig. 1 controls the temperature of a low temperature shift reactor and makes beneficial use of the heat of reaction to generate steam that is employed to produce hydrogen-rich gases containing low concentrations of carbon monoxide from hydrocarbon feedstocks.

A reactant mixture at 1 consists of hydrocarbon feedstock 32 and steam 11. The mixture is preheated in an exchanger 2 and introduced into a tubular catalytic reactor 3 that is contained within a combustion chamber 4. The tubular catalytic reactor typically contains a supported Ni catalyst and is

commonly referred to in the industry as a steam reformer. Fuel 5 and air 6 are combusted in the chamber 4 to heat the reactant mixture so as to produce a hydrogen-rich stream 7 containing carbon monoxide concentrations typically ranging from 5% to 15%.

Combustion products 8 from the combustion chamber pass through a flue gas heat exchange coil 9 that is contained within a waste heat stem generator 10, wherein the combustion products are cooled and stem 11 is generated. The cooled combustion products 13 are further cooled by exchanging heat in a feed water exchanger 14 that produces heated water 15 that is supplied to the waste heat steam generator 10.

The hydrogen-rich stream 7 from the tubular catalytic reactor 3 is cooled in an exchanger 2 to a temperature typically in the range of 400°F-550°F whereupon the cooled steam 18 is introduced into a fixed-bed catalytic reactor 19 shown as surrounding steam generator 10, to effect a water gas shift reaction that converts a portion of the carbon monoxide to hydrogen and carbon dioxide by reaction with steam. The catalyst bed reactor typically contains a supported Cu/Zn catalyst and is commonly known in the industry as a low temperature shift reactor. The walls 20 of the low temperature shift reactor are in thermal communication with boiling water contained in the waste

heat steam generator. The heat released in the low temperature shift reactor is thus beneficially recovered to generate steam. The carbon monoxide concentration of the process gas 21 exiting the low temperature shift reactor is typically less than 0.5%.

The products from the low temperature shift reactor are cooled in a process exchange coil 22 to condense moisture. The condensed moisture 40a is separated in a separator vessel 28 and is pumped at 40b to heat exchanger 14 using pump 41. Make-up water may be added as at 40c. The cooled process gas 29 is mixed with a small quantity of air 30 and is sent to the selective oxidizer reactor 31. The required quantity of ambient air that must be introduced into the process gas depends on the specific performance of the catalyst but typically ranges from 2 to 4 times the quantity necessary to provide a stoichiometric quantity oxygen for the complete oxidation of carbon monoxide.

The process gas 32 exiting the selective oxidizer typically contains less than 10 ppm carbon monoxide and is available for use, for instance, in fuel cells having a low tolerance to carbon monoxide. A fuel cell is indicated at 100.

EXPERIMENTAL DATA

Tests were conducted to measure the conversion of carbon monoxide contained in a simulated reformate gas stream that was passed over a commercial Cu/Zn catalyst. The Cu/Zn catalyst was contained within tubular conduit walls that were in thermal communication with a boiling water fluid. The objective of the tests was to quantify the relation between CO conversion and catalyst gas hourly space velocity (GHSV) as a function of the temperature of the boiling water fluid.

The experimental test system is shown in Fig. 2. The Cu/Zn catalyst 80 was packed in a ¾ inch tube 81 that was surrounded by a metallic water jacket 82. Steam 83 generated from the water jacket was sent to a condenser 84 and the condensed liquid 85 was returned to the water jacket by natural circulation. An electrical heater 86 was placed on the outer surface of the jacket and a temperature controller 87 was used to control the heat input in order to maintain the water jacket at the desired temperature. A simulated reformate gas mixture 88 was heated in a pre-heater 89 to a temperature approximately equal to the boiling water fluid temperature before the gases entered the catalyst bed. The product gases 90 at the exit of the

catalyst bed were cooled, condensed and sent to a gas chromatograph for compositional analysis. Table 2 summarizes the results for tests conducted using the experimental apparatus.

Table 2. Carbon Monoxide exit concentration as a function of boiler temperature and catalyst space velocity

Simulated Reformate Composition at Inlet to Reactor:

Component	Wet Gas Vol.%	Dry Gas Vol.%
CO	5.70	10.0
CO_2	8.55	15.0
H_2	39.90	70.0
CH_4	2.85	5.0
H_2O	43.00	0.0
Total	100.00	100.0

Dry Gas GHSV (hr-1)	Boiler Temperature°F	Carbon Monoxide Exit Concentration (Vol.% dry gas)
1930	340	3.67
762	340	0.62
2950	385	2.84
2440	385	2.08
1930	385	1.70
1425	385	0.90
1930	398	1.08
1425	398	0.55

In order to maximize hydrogen yield and minimize the quantity of carbon monoxide that must be converted in the selective oxidizer, it is generally desirable to achieve less than 1.0 vol.% CO in the dry gas at the exit of the shift reactor. It is desirable to operate at the maximum space velocity in order to minimize the size of the catalyst bed.

The results show that when the boiler is maintained at temperatures above about 385°F, the CO concentration is less than about 1.0 vol.% (dry gas) for space velocities less than or equal to about 1500hr-1. Higher space velocities can be achieved for even higher boiler temperatures, however this necessitates operation at pressures above 250 psig,

which may not be desirable for many PEM fuel cell applications.

The results indicate that there is an optimum temperature range of operation for the boiler that achieves the objects of minimum catalyst volume, moderate boiler pressure and low CO concentration at the shift reactor exit. This optimum temperature range is approximately 385°F to 400°F.

THERMALLY-INTEGRATED LOW TEMPERATURE WATER-GAS SHIFT REACTOR APPARATUS

Apparatus shown in Figs. 3a and 3b controls the temperature of a low temperature shift reactor and makes beneficial use of the heat of reaction to generate steam.

In the preferred embodiment, the apparatus comprises an annular catalyst zone 50 within vessel 50a that is concentrically disposed around an inner steam generator zone 51 within vessel 51a. The catalyst zone contains a lower temperature shift catalyst, typically comprising a catalytically active mixture of copper and zinc metals, that promote the water-gas shift reaction at temperatures typically in the range of 370°F to 500°F. The catalyst zone is defined by the annular space between an outer wall 52 and an inner wall 53.

The steam generator zone is defined in the space inside of the inner wall, a top wall 65, and a tube sheet 66.

Reformate containing carbon monoxide enters an inlet conduit 54 that is in communication with the annular catalyst zone. The reformate passes through the catalyst zone, releasing heat due to the exothermic nature of the water-gas shift reaction. The reformate exits from the catalyst zone through an exit conduit 55.

The inner wall 53 is in thermal contact with the reformate gases flowing through the catalyst zone and boiling water 56 contained in the steam generator zone, the catalyst zone shown as surrounding the steam generation zone. This results in a transfer of heat from the catalyst zone to the steam generator zone in order to maintain the catalyst zone within a defined temperature range that is optimum for conducting the low temperature shift reaction.

The steam generator zone contains a heat transfer conduit 57, or a multitude of heat transfer conduits, that is immersed within the boiling water. The heat transfer conduit conveys hot combustion products, such as from the combustion section of a steam reformer, to transfer heat to the boiling water for the purpose of generating steam. The hot combustion products enter an inlet plenum 58 through an

inlet conduit 59. The inlet plenum is in communication with the inlet portion 60 of the heat transfer conduit. The hot combustion products exit the heat transfer conduit to an exit plenum 61 that is connected to an exit conduit 62. Steam that is generated from the heat transferred to the steam generator zone through the inner wall 53 and the heat transfer conduit 57 exits the steam generator zone through an exit conduit 68. Water is fed to the steam generator zone through an inlet conduit 69 to maintain the boiling water at a desired level that is sufficient to efficiently remove heat from the inner wall and the heat transfer conduit.

The annular catalyst zone 50 may include means to enhance heat transfer between the catalyst bed and the inner wall 53 such as the use of longitudinal fins 70 that are attached to the inner wall and extend a distance into the annular catalyst space. Other heat transfer enhancements such as metal packing within the annular catalyst zone, or active catalyst that are incorporated into highly thermally conductive monoliths, may be beneficially used in the invention.

EXAMPLE

A pipe wall having an outside diameter of 5.5 inches defines a waste heat steam generator zone. The pipe wall is in thermal communication with an annular

catalyst zone that is concentrically disposed around the waste heat steam generator zone. The space between the 5.5 inch diameter pipe and an outer wall having an 8 inch inside diameter define the annular catalyst zone. The annular catalyst zone is packed with 4.8 mm x 2.8 mm low temperature shift catalyst containing principally a mixture of Cu and Zn supported on a ceramic carrier. The packed height of the annular catalyst zone is approximately 12 inches and corresponds to the approximate height of the water contained within the waste heat steam generator zone. The total catalyst volume is approximately 0.184 cubic feet.

The waste heat steam generator zone also contains heat transfer conduits that are immersed within the boiling water. The heat transfer conduits consist of 5/8 inch diameter tubes that are formed in a u-tube arrangement having inlet means and exit means that are attached to a single tube sheet. Hot combustion products from a steam reformer are directed to the inlet means of the heat transfer tubes. Cooled combustion products are exhausted from the exit means of the heat transfer tubes.

The waste heat steam generator zone is operated at a controlled pressure using a back-pressure regulator that is located on the steam exit conduit.

The back-pressure regulator is set at a pressure of 220 psia corresponding to a saturated water temperature of 390°F.

Reformate from a tubular catalytic reactor is cooled to approximately 510°F and is introduced into the top of the annular catalyst zone. The reformate exits the bottom of the annular catalyst zone at a temperature of approximately 430°F. The composition and flow rate of the reformate at the inlet and exit of the annular catalyst zone is shown in Table 3.

Table 3. Gas composition from thermally-integrated low temperature water-gas shift reactor

	Gas Composition (Volume %)	
Component	Reformate Inlet	Reformate Exit
CH4	2.1	2.1
CO	8.1	0.8
CO_2	7.5	14.7
H_2	52.6	59.8
H_2O	29.7	22.5
Total	100.00	100.0
Total Flow, SCFH	280	280
Temperature, °F	510	430
Pressure, psia	16.7	16.7

ALTERNATIVE EMBODIMENT OF THERMALLY-INTEGRATED LOW TEMPERATURE WATER-GAW SHIFT REACTOR APPARATUS

Fig. 4 depicts an alternative embodiment of the thermally-integrated low temperature water-gas shift reactor apparatus. A helical coil 75 is welded to the surfaces of the inner wall 53 and outer wall 52 that define the annular catalyst space. The catalyst is packed within the volumes defined by the helical coil and the inner and outer walls. The gases entering the inlet conduit 54 are directed in a helical manner around the annular space by virtue of the confining helical coil. This increases the velocity of the gases within the catalyst bed for a given space velocity in order to enhance the rate of heat transfer to and from the bed. The helical coil also serves as an extended heat transfer surface to enhance the rate of heat transfer to and from the inner wall of the annular space. Elements the same as those of Fig. 3a bear the same identifying numbers.

WE CLAIM:

1. A thermally-integrated low temperature water-gas shift reactor for converting carbon monoxide in the presence of steam to form carbon dioxide and water comprising, in combination,

a) a waste-heat recovery steam generator for the beneficial recovery of exothermic reaction heat to generate steam that is used in a process for the conversion of hydrocarbon feedstock into hydrogen-rich gases,

b) an outer region extending about said waste-heat steam generator,

c) a catalyst bed located within said outer region, and through which reformate gases flow,

d) the outer region being in heat transfer communication with the steam generator to maintain the catalyst bed within a predetermined temperature range for operation of a low temperature shift reaction.

2. The combination of claim 1 wherein the waste heat steam generator operates at temperatures in one of the following ranges: 360°F to 450°F, and of 385°F to 400°F, that is optimum for conducting the low temperature water-gas shift reaction.

3. The combination of claim 1 wherein a Cu/Zn catalyst is contained in an annular space defined by said outer region, and having an inner wall that is in thermal contact with a boiling water fluid in said generator.

4. The combination of claim 3 wherein the boiling water fluid is located to transfer heat to the catalyst bed to heat the bed during start-up.

5. The combination of claim 3 wherein the catalyst bed is located to transfer heat to the boiling water fluid during normal operation to generate steam.

6. The combination of claim 1 including longitudinal heat transfer fins attached to an inside wall of the annulus and projecting in said bed to enhance the rate of heat transfer to and from the catalyst bed.

7. The combination of claim 1 including a helical coil adjacent the inner and outer walls for defining annular space at said annulus to conduct and increase the velocity of the process gases as they flow through the catalyst bed and to enhance the rate of heat transfer to and from the catalyst bed.

8. The combination of claim 1 wherein the catalyst bed is maintained in one of the following ranges: between 370°F and 550°F, and between 400°F and 450°F.

9. The combination of claim 3 wherein the annular space is typically between 1 and 2 inches wide to minimize temperature differentials between the outside and inside walls defined by the annular space.

10. The combination of claim 3 wherein the gas has hourly space velocity in the range of 500hr-1 to 2000hr-1.

11. The combination of claim 1 wherein the waste heat stem generator contains one or more heat transfer conduits that transfer heat from hot combustion products to a boiling water fluid for the purpose of generating steam.

12. The combination of claim 1 wherein the steam generator includes an upright vessel, said outer region having an upper level inlet flowing reformate into the catalyst bed, the reformate containing carbon monoxide, and said region having a lower level outlet, a catalyst bed located between said upper and lower levels, a heat transfer conduit or conduits extending within said vessel and immersed within boiling water contained in said vessel inwardly of said catalyst bed, said conduit or conduits receiving hot products of combustion from a combustion process, for transfer of heat to the boiling water, for generating steam exiting from said vessel.

13. The combination of claim 1 wherein the catalyst bed extends helically about said generator.

14. The combination of claim 1 including a helical coil in said outer region and extending about said generator, to direct said reformate gases helically and through said catalyst bed, to enhance heat transfer via said bed.

ABSTRACT OF THE DISCLOSURE

A thermally-integrated lower temperature water-gas shift reactor apparatus for converting carbon monoxide in the presence steam comprises a catalyst bed that is disposed within an outer region surrounding a waste heat recovery steam generator operating at a selected pressure corresponding to the optimum temperature for conducting the catalytic water-gas shift reaction and a process for useful recovery of the exothermic heat of reaction to generate steam that is used in a process for the conversion of hydrocarbon feedstock into useful gases such as hydrogen.

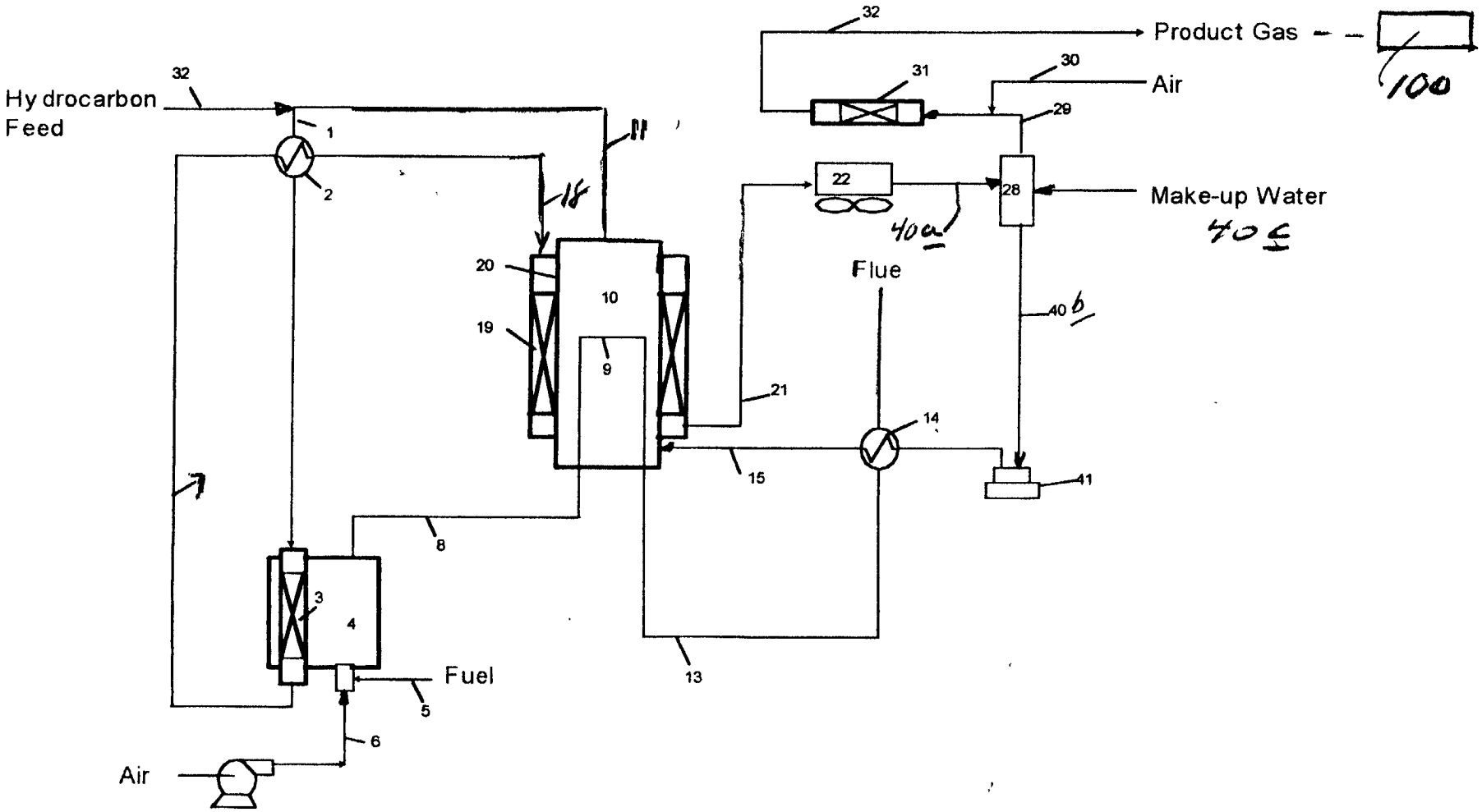

Fig 1

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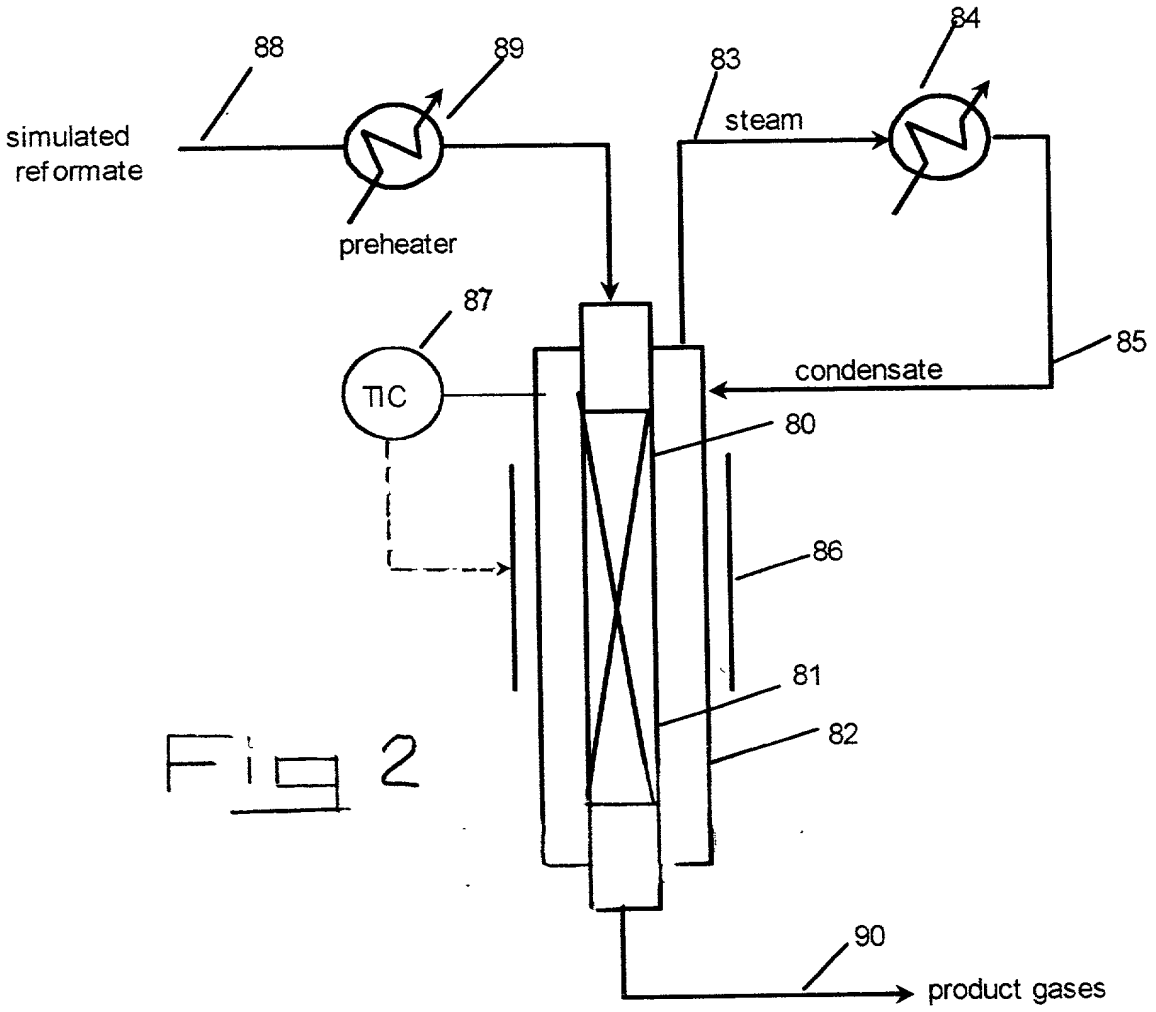
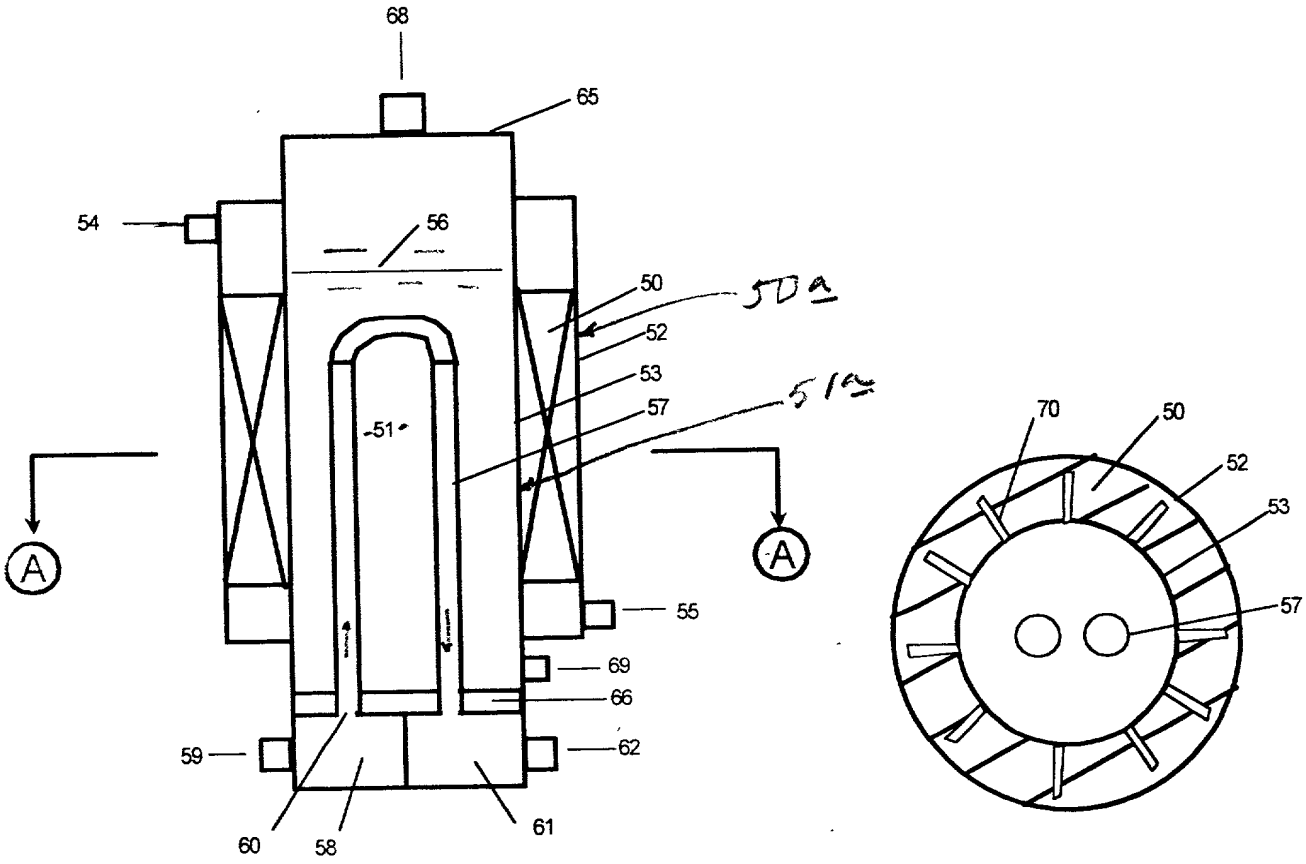


Fig 2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SIDE VIEW
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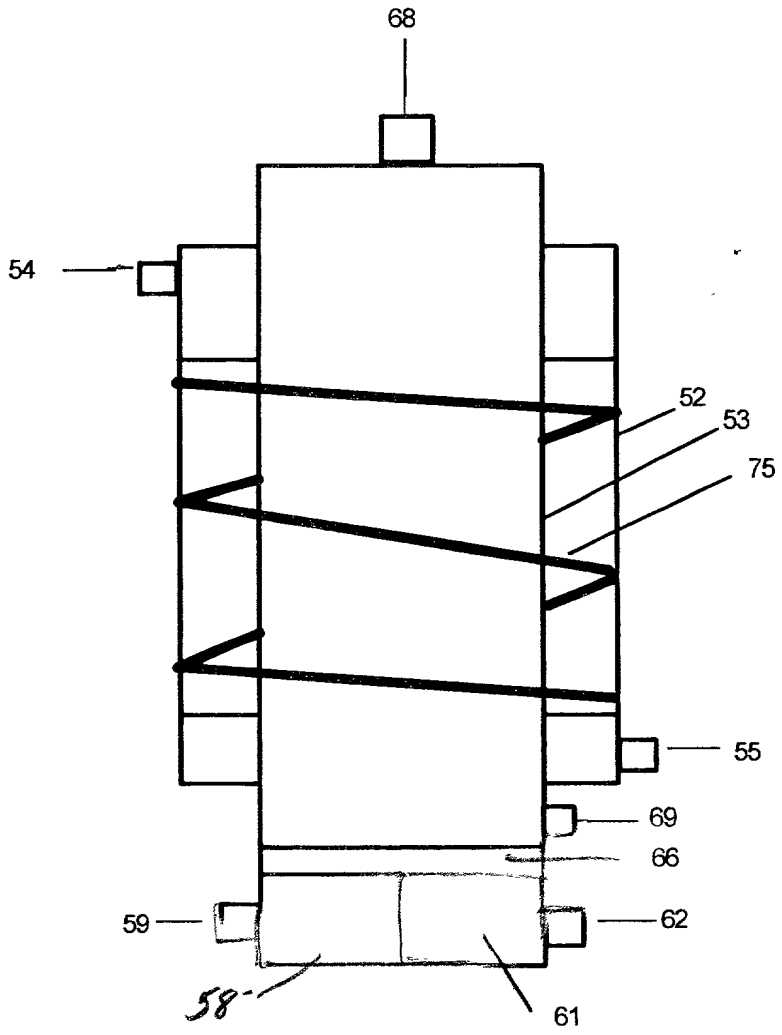
SECTION A
(a)
(b)

Fig 3



SIDE VIEW

Fig 4

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION (37 CFR 1.63)

☒ Declaration Submitted with Initial Filing **OR** ☐ Declaration Submitted after Initial Filing (surcharge (37 CFR 1.16 (e)) required)

Attorney Docket Number	12,150
First Named Inventor	David W. Warren
COMPLETE IF KNOWN	
Application Number	/
Filing Date	
Group Art Unit	
Examiner Name	

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

THERMALLY-INTEGRATED LOW TEMPERATURE WATER-GAS SHIFT REACTOR APPARATUS AND PROCESS

(Title of the Invention)

the specification of which
☒ is attached hereto
OR
☐ was filed on (MM/DD/YYYY) [ ] as United States Application Number or PCT International Application Number [ ] and was amended on (MM/DD/YYYY) [ ] (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. 119(a)-(d) or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES	NO
			☐	☐	☐
			☐	☐	☐
			☐	☐	☐
			☐	☐	☐

☐ Additional foreign application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto:

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	
		☐ Additional provisional application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

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DECLARATION — Utility or Design Patent Application

I hereby claim the benefit under 35 U.S.C. 120 of any United States application(s), or 365(c) of any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application or PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number *(if applicable)*

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority data sheet PTO/SB/02B attached hereto.

As a named inventor, I hereby appoint the following registered practitioner(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith: ☐ Customer Number [ ] → *Place Customer Number Bar Code Label here*
OR
☒ Registered practitioner(s) name/registration number listed below

Name	Registration Number	Name	Registration Number
William W. Haefliger	17,120		

☐ Additional registered practitioner(s) named on supplemental Registered Practitioner Information sheet PTO/SB/02C attached hereto.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])	Family Name or Surname
David W.	Warren

Inventor's Signature	David W. Warren					**Date**	11/1/00
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Post Office Address							
City	Van Nuys	**State**	CA	**ZIP**	91401	**Country**	U.S.A.

☒ Additional inventors are being named on the ____supplemental Additional Inventor(s) sheet(s) PTO/SB/02A attached hereto

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DECLARATION

ADDITIONAL INVENTOR(S)
Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

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Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

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Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name (first and middle [if any])				Family Name or Surname	
Inventor's Signature				**Date**	
Residence: City		**State**		**Country**	**Citizenship**
Post Office Address					
Post Office Address					
City		**State**	**ZIP**	**Country**	